

PATENT
TS1313 (US)
CWS:EM

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE BOARD OF APPEALS AND INTERFERENCES

In re application of)
CAROLUS M. A. M. MESTERS and)
RONALD J. SCHOONEBEEK)
Serial No. 10/738,332) Group Art Unit: 1764
Filed December 17, 2003) Examiner: John Christopher Douglas
PROCESS FOR THE CATALYTIC) March 26, 2007
SELECTIVE OXIDATION OF SULFUR)
COMPOUNDS)

)

COMMISSIONER FOR PATENTS
P. O. Box 1450
Alexandria, VA 22313-1450

Sir:

APPEAL BRIEF

REAL PARTY IN INTEREST

The real party in interest is the assignee of record, Shell Oil Company.

RELATED APPEALS AND INTERFERENCES

There are no other prior and pending appeals, interferences or judicial proceedings known to the appellant which may be related to, directly affect or be directly affected by or have a bearing on the Board's decision in the pending appeal.

STATUS OF CLAIMS

The claims 1-2, 9, 11-21, 24-26, 30, and 33-44 are finally rejected.

The claims 3-8, 10, 22-23, 27-29, and 31-32 are canceled.

The claims on appeal are 1-2, 9, 11-21, 24-26, 30, and 33-44.

STATUS OF AMENDMENTS

No amendment has been filed subsequent to the Examiner's final rejection.

The claims on appeal are those presented in the Applicant's amendment mailed on 9 August 2006 in response to the Examiner's non-final office action mailed on 24 April 2006.

SUMMARY OF CLAIMED SUBJECT MATTER

Claim 1

A process for the catalytic selective oxidation of a sulfur compound contained in a hydrocarbonaceous feedstock to sulfur dioxide [paragraphs 0001, 0006, 0010-0012 of the published specification], wherein the process comprises: contacting a gaseous feed mixture of the hydrocarbonaceous feedstock [0007, 0009-0010, 0046, 0048, 0053, 0056, 0060], of which the sulfur compound is selected from the group consisting of hydrocarbon sulfide, mercaptans, disulfides and heterocyclic sulfur compounds [0013], and a molecular-oxygen containing gas [0007, 0012, 0022] with a catalyst at a temperature of at most 500 °C [0007-0009, 0012, 0019], wherein said catalyst comprises a catalytically active group VIII noble metal selected from the group consisting of platinum, rhodium, iridium and combinations of two or more thereof [0007, 0009-0010, 0012, 0017-0018, 0031, 0035, 0037, 0041, 0051, 0058] at a concentration in the range of from 0.02 to 10% by weight, based on the total weight of the catalyst [0018, 0031, 0035, 0037, 0041, 0051, 0058], supported on a catalyst carrier comprising stabilized or partially stabilized zirconia [0015, 0031, 0035, 0037, 0041, 0051, 0058], wherein said feed mixture has an oxygen-to-carbon ratio of below 0.15 [0009, 0012, 0024].

Claim 21

A process for the catalytic selective oxidation of hydrogen sulfide contained in a methane or natural gas feedstock to sulfur dioxide [0001, 0006, 0010-0012], wherein the process

comprises: contacting a gaseous feed mixture of the methane or natural gas feedstock [0007, 0009-0010, 0046, 0048, 0053, 0056, 0060] and a molecular-oxygen containing gas [0007, 0012, 0022], wherein the gaseous feed mixture comprises up to 10% v/v hydrogen sulfide [0025], with a catalyst at a temperature of at most 500 °C [0007-0009, 0012, 0019], wherein said catalyst comprises a catalytically active group VIII noble metal selected from the group consisting of platinum, rhodium, iridium and combinations of two or more thereof [0007, 0009-0010, 0012, 0017-0018, 0031, 0035, 0037, 0041, 0051, 0058] at a concentration in the range of from 0.02 to 10% by weight, based on the total weight of the catalyst [0018, 0031, 0035, 0037, 0041, 0051, 0058], supported on a refractory oxide comprising stabilized or partially stabilized zirconia [0015, 0031, 0035, 0037, 0041, 0051, 0058], wherein said feed mixture has an oxygen-to-carbon ratio of below 0.15 [0009, 0012, 0024].

Claim 25

A process for the desulfurization of a hydrocarbonaceous feedstock, wherein the process comprises: contacting a gaseous feed mixture of the hydrocarbonaceous feedstock [0007, 0009-0010, 0046, 0048, 0053, 0056, 0060], which contains a sulfur compound is selected from the group consisting of hydrocarbon sulfide, mercaptans, disulfides and heterocyclic sulfur compounds [0013], and a molecular-oxygen containing gas [0007, 0012, 0022] with a catalyst at a temperature of at most 500 °C [0007-0009, 0012, 0019], wherein said catalyst comprises a catalytically active group VIII noble metal selected from the group consisting of platinum, rhodium, iridium and combinations of two or more thereof [0007, 0009-0010, 0012, 0017-0018, 0031, 0035, 0037, 0041, 0051, 0058] at a concentration in the range of from 0.02 to 10% by weight, based on the total weight of the catalyst [0018, 0031, 0035, 0037, 0041, 0051, 0058], supported on a refractory oxide comprising partially stabilized or stabilized zirconia [0015, 0031, 0035, 0037, 0041, 0051, 0058], wherein said gaseous feed mixture has an oxygen-to-carbon ratio of below 0.15 [0009, 0012, 0024], thereby selectively oxidizing the sulfur compounds in the hydrocarbonaceous feedstock to sulfur dioxide [0001, 0006, 0010-0011, 0013]; and removing the thus-formed sulfur dioxide from the hydrocarbonaceous feedstock [0026-0028].

GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

Whether claims 1-2, 9, 11-21, 24-26, 30, and 33-44 are unpatentable under 35 U.S.C §103 over Frame (US 3,978,137) in view of Yoo (US 3,945,914) and Mahadev (WO 92/20621).

ARGUMENT

The §103 rejection of claims 1-2, 9, 11-21, 24-26, 30, and 33-44 over Frame in view of Yoo and Mahadev.

The primary reference of Frame teaches a process for the oxidation of sulfur compounds using a catalyst system that comprises both a Group VIIB (e.g. manganese and rhenium) metal phthalocyanine and a Group VIII (e.g. cobalt, iron, nickel, palladium, rhodium, ruthenium, osmium, iridium, and platinum) metal phthalocyanine.

The secondary reference of Yoo teaches a two-step process involving a first step of oxidation of sulfur contained in a hydrocarbon material in the presence of an oxidant and, preferably, a metal-containing catalyst, see e.g. column 2, lines 46-63, followed by contacting the oxidized sulfur-containing hydrocarbon material with a metal-containing compound under certain temperature conditions so as to form a metal-sulfur-containing compound. See e.g. column 1, lines 50-61; and column 7, lines 10-30. The preferred catalyst metals for the oxidation catalyst are Group IVB, Group VB, and Group VIB metals. See column 4, lines 7-10. The Examiner uses the Yoo patent to supply teachings concerning the amount of oxygen that may be in the hydrocarbon feed.

The third reference of Mahadev discloses a process for the removal of hydrogen sulfide and other sulfur compounds from sour natural gas and other fluid streams and the conversion of the sulfur therein to elemental sulfur. See e.g. page 1, lines 9-19. The Examiner uses the Mahadev publication to supply teachings concerning concentration ranges for sulfur compounds contained in a natural gas feedstream.

The Applicant respectfully submits that no modification of the Frame reference can possibly result in providing a process as claimed by the Applicant. Moreover, the references cited by the Examiner are not properly combinable.

As summarized above, Frame teaches the use of an oxidation catalyst system comprising both a Group VIIB metal phthalocyanine and a Group VIII metal phthalocyanine. The compounds of the catalyst system of the Frame patent are not similar in any way to the oxidation catalyst of the Applicant's claimed invention. The Applicant's claimed oxidation catalyst comprises a Group VIII noble metal on an at least a partially stabilized zirconia support. The two catalysts are hugely different. One of the differences, for example, is the requirement that the

Frame catalyst includes a Group VIIB metal (Mn or Re). Another difference is the requirement by Frame that the catalytic components of its catalyst be metal phthalocyanine compounds.

The Frame catalyst is so different from the Applicant's claimed catalyst that there is no way that it may be modified by the application of the secondary and tertiary references to give the Applicant's claimed catalyst. Furthermore, the process of the Frame patent is, as well, hugely different from the Applicant's claimed process. For instance, the Frame process requires its treating of a sulfur-containing gas to be in the presence of a medium having a pH of from 8 to 14.

The Examiner's response to the Applicants' comments concerning the differences between the catalyst of the Frame patent and the catalyst as recited in the Applicants' claims is that the noted differences are not features that are recited in the rejected claims. However, the Examiner's position is not completely correct; because, there are both features explicitly recited in the claimed catalyst that are not mentioned in the Frame patent and there are features indicated in the Frame patent as being necessary but which are not recited in the Applicants' claims. For instance, the claimed catalyst is a noble metal (platinum, rhodium and iridium) supported on a stabilized or partially stabilized zirconia. The Frame patent does not teach this type of catalyst. It certainly says nothing about the use of stabilized or partially stabilized zirconia as a carrier, and the catalytic compounds of the Frame patent are necessarily phthalocyanine compounds. Specifically, the phthalocyanine compounds are selected from Group VIIB metal phthalocyanines and Group VIII metal phthalocyanines.

Additionally, the Frame patent teaches that its catalyst system must include both the Group VIIB metal and Group VIII metal phthalocyanine compounds. The Applicants' claimed catalyst, on the other hand, does not necessarily include a Group VIIB metal, and the form of the Group VIII metal is not that of a phthalocyanine. As may be seen from the Applicants' specification, the catalyst is stabilized or partially stabilized zirconia that has been impregnated with the Group VIII metal (usually as a salt) and subsequently dried and calcined.

Another feature not taught by the Frame patent, as admitted by the Examiner in his office action, is that of the concentration of noble metal included in the catalyst. In fact, the Frame patent is not actually able to present this type of concentration information; since, the focus of the Frame patent is primarily on the use of its catalyst system that includes the two phthalocyanine metal compounds that are in liquid form with the acceptable amounts of each component being expressed in terms of relative amounts to each other. While the Frame patent

indicates a range for the weight percent of the catalyst system relative to the entire reaction system, it is not really clear what this actually means. But, in any event, there is no clear teaching in the Frame patent as to how much of the phthalocyanine metal compounds is to be on a carrier.

In addition to the above-noted differences of the claimed invention over the teachings of the primary reference, the teachings of the secondary references of Yoo and Mahadev are not properly combinable with those of the primary reference of Frame due to the processes and catalysts that are taught in the secondary references being substantially different from those taught by the primary reference. Furthermore, the primary and secondary references are not properly combinable because there are no teachings within them to suggest their combination. But, even if they were to be combinable, any combination fails to disclose all the limitations and features of the Applicants' claimed process. The Examiner even admits that the references do not teach the Applicants' claimed noble metal concentration range, and, in any event, none of the references teaches a catalyst that includes stabilized or partially stabilized zirconia for use as a carrier for a noble metal. Moreover, the claimed catalyst does not necessarily include a Group VIIB metal phthalocyanine compound as is required by the Frame reference.

CONCLUSION

In view of the above remarks, it is submitted that the claims on appeal in this case are patentable. Therefore, the reversal of the Examiner's final rejection thereof is respectfully requested.

Respectfully submitted,

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CLAIMS APPENDIX

1. (Previously Presented) A process for the catalytic selective oxidation of a sulfur compound contained in a hydrocarbonaceous feedstock to sulfur dioxide, wherein the process comprises: contacting a gaseous feed mixture of the hydrocarbonaceous feedstock, of which the sulfur compound is selected from the group consisting of hydrogen sulfide, mercaptans, disulfides, and heterocyclic sulfur compounds, and a molecular-oxygen containing gas with a catalyst at a temperature of at most 500 °C, wherein said catalyst comprises a catalytically active group VIII noble metal selected from the group consisting of platinum, rhodium, iridium and combinations of two or more thereof at a concentration in the range of from 0.02 to 10% by weight, based on the total weight of the catalyst, supported on a catalyst carrier comprising stabilized or partially stabilized zirconia, wherein said feed mixture has an oxygen-to-carbon ratio of below 0.15.
2. (Original) The process of claim 1 wherein the oxygen-to-carbon ratio of the feed mixture is below 0.10.

Claims 3-8 (Canceled).

9. (Previously Presented) The process of claim 2 wherein the temperature is maintained in the range of from 200 to 500°C.

Claim 10 (Canceled).

11. (Original) The process of claim 1 wherein the feed mixture is contacted with the catalyst at a pressure in the range of from 1 to 10 bar (absolute).
12. (Original) The process of claim 11 wherein the feed mixture is contacted with the catalyst at a pressure in the range of from 1 to 5 bar (absolute).
13. (Original) The process of claim 1 wherein the feed mixture is contacted with the catalyst at ambient pressure.
14. (Original) The process of claim 1 wherein the hydrocarbonaceous feedstock is a gaseous hydrocarbonaceous feedstock.
15. (Original) The process of claim 14 wherein the hydrocarbonaceous feedstock is methane or natural gas.

16. (Original) The process of claim 14 wherein the hydrocarbonaceous feedstock comprises hydrogen sulfide in a concentration of at most 10% v/v.
17. (Original) The process of claim 16 wherein the hydrocarbonaceous feedstock comprises hydrogen sulfide in a concentration of at most 5% v/v.
18. (Original) The process of claim 15 wherein the hydrocarbonaceous feedstock comprises hydrogen sulfide in a concentration of at most 10% v/v.
19. (Original) The process of claim 18 wherein the hydrocarbonaceous feedstock comprises hydrogen sulfide in a concentration of at most 5% v/v.
20. (Original) The process of claim 1 wherein the feedstock is a liquid hydrocarbonaceous feedstock containing at most 1000 ppmw sulfur.
21. (Previously Presented) A process for the catalytic selective oxidation of hydrogen sulfide contained in a methane or natural gas feedstock to sulfur dioxide, wherein the process comprises: contacting a gaseous feed mixture of the methane or natural gas feedstock and a molecular-oxygen containing gas, wherein the gaseous feed mixture comprises up to 10% v/v hydrogen sulfide, with a catalyst at a temperature of at most 500 °C, wherein said catalyst comprises a catalytically active group VIII noble metal selected from the group consisting of platinum, rhodium, iridium and combinations of two or more thereof at a concentration in the range of from 0.02 to 10% by weight, based on the total weight of the catalyst, supported on a refractory oxide comprising stabilized or partially stabilized zirconia, wherein said feed mixture has an oxygen-to-carbon ratio of below 0.15.

Claims 22-23 (Canceled).

24. (Previously Presented) The process of claim 21 wherein the hydrocarbonaceous feedstock comprises hydrogen sulfide in a concentration of at most 5% v/v.
25. (Previously Presented) A process for the desulfurization of a hydrocarbonaceous feedstock, wherein the process comprises: contacting a gaseous feed mixture of the hydrocarbonaceous feedstock, which contains a sulfur compound selected from the group consisting of hydrogen sulfide, mercaptans, disulfides, and heterocyclic sulfur compounds, and a molecular-oxygen containing gas with a catalyst at a temperature of at most 500 °C, wherein said catalyst comprises a catalytically active group VIII noble metal selected from the group consisting of platinum, rhodium, iridium and combinations of

two or more thereof at a concentration in the range of from 0.02 to 10% by weight, based on the total weight of the catalyst, supported on a refractory oxide catalyst carrier comprising partially stabilized or stabilized zirconia, wherein said gaseous feed mixture has an oxygen-to-carbon ratio of below 0.15, thereby selectively oxidizing the sulfur compounds in the hydrocarbonaceous feedstock to sulfur dioxide; and removing the thus-formed sulfur dioxide from the hydrocarbonaceous feedstock.

26. (Previously Presented) The process of claim 25 wherein the oxygen-to-carbon ratio of the gaseous feed mixture is below 0.10.

Claims 27-29 (Canceled).

30. (Previously Presented) The process of claim 26 wherein the oxygen-to-carbon ratio of the gaseous feed mixture is below 0.10.

Claims 31-32 (Canceled).

33. (Original) The process of claim 25 wherein the temperature is maintained in the range of from 200 to 500°C.

34. (Original) The process of claim 25 wherein the temperature is maintained in the range of from 200 to 300°C.

35. (Previously Presented) The process of claim 25 wherein the gaseous feed mixture is contacted with the catalyst at a pressure in the range of from 1 to 10 bar (absolute).

36. (Previously Presented) The process of claim 35 wherein the gaseous feed mixture is contacted with the catalyst at a pressure in the range of from 1 to 5 bar (absolute).

37. (Previously Presented) The process of claim 25 wherein the gaseous feed mixture is contacted with the catalyst at ambient pressure.

38. (Original) The process of claim 25 wherein the hydrocarbonaceous feedstock is a gaseous hydrocarbonaceous feedstock.

39. (Original) The process of claim 38 wherein the hydrocarbonaceous feedstock is methane or natural gas.

40. (Original) The process of claim 38 wherein the hydrocarbonaceous feedstock comprises hydrogen sulfide in a concentration of at most 10% v/v.

41. (Original) The process of claim 40 wherein the hydrocarbonaceous feedstock comprises hydrogen sulfide in a concentration of at most 5% v/v.
42. (Original) The process of claim 39 wherein the hydrocarbonaceous feedstock comprises hydrogen sulfide in a concentration of at most 10% v/v.
43. (Original) The process of claim 42 wherein the hydrocarbonaceous feedstock comprises hydrogen sulfide in a concentration of at most 5% v/v.
44. (Original) The process of claim 25 wherein the feedstock is a liquid hydrocarbonaceous feedstock containing at most 1000 ppmw sulfur.

EVIDENCE APPENDIX

None.

RELATED PROCEEDINGS APPENDIX

None.